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Revision of the Transuranus PUREDI Model

Technical report for the JRC-ITU Action No. 52201 – Safety of Nuclear Fuels and Fuel cycles

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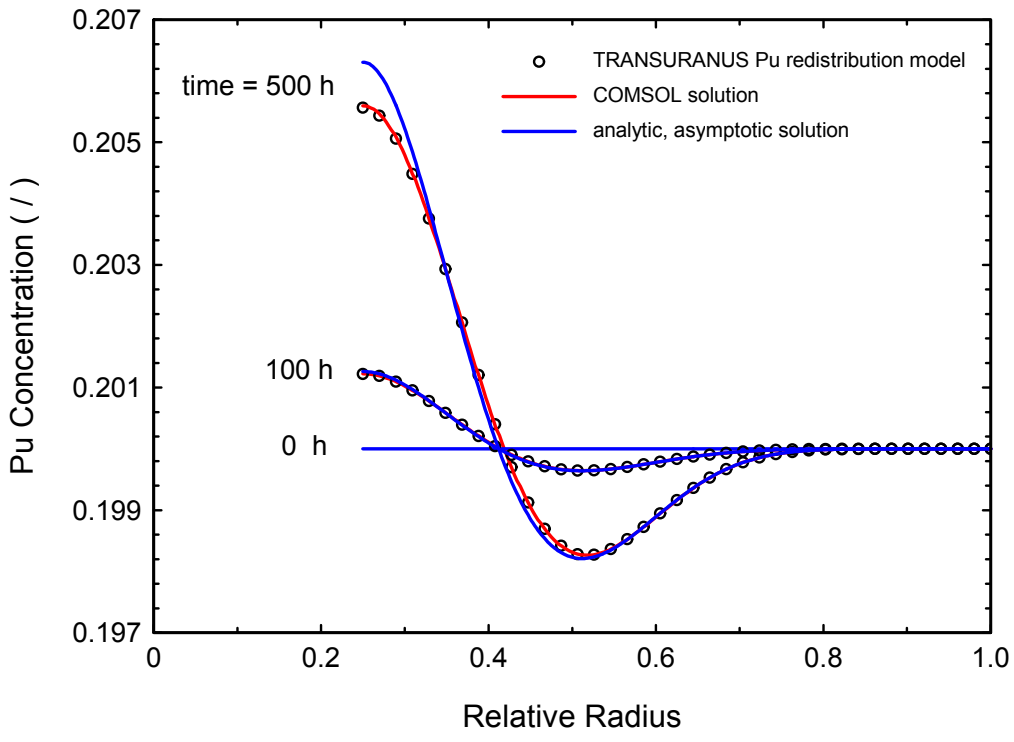

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1. Introduction

The use of appropriate models for redistribution is an important added value for fuel performance codes for the description of the actinides transport and evolution across the fuel pellet. Actually, modelling of the plutonium radial distribution is a key issue for design purposes of FBR fuel pins, since Pu build-up in the central part of the fuel pellet during irradiation can impose significant constraints on the allowed maximum fuel temperature and therefore on the linear heat rating [1]. Plutonium evolves and migrates during irradiation mainly because of burn-up (fissions and transmutations) and temperature gradient (diffusion), respectively. The burn-up evolution is calculated in Transuranus by means of TUBRNP (recently extended for FBRs) for each different plutonium isotope according to neutronic features of the reactor. The PUREDI model calculates the diffusion of the total plutonium concentration, but the two models, TUBRNP and PUREDI, are not coupled. For this reason, we propose a revision of PUREDI so that, at the end of a time step, it returns the "redistributed" concentrations of the different isotopes to be used by TUBRNP during the next time step. We identified two possibilities to perform the above mentioned coupling: 1) the diffusion equation is solved separately for each different plutonium isotope; 2) the "redistributed" isotope concentrations are calculated from the total plutonium concentration by means of a simplified "splitting" formula. The two options are discussed in the report, and the second one is chosen for implementation, since it allows to safe computational time, while maintaining the same accuracy when experimental uncertainties are taken into account.

The report is structured as follows: in the second section the description of the stand-alone PUREDI model and the assessment of the numerical solution will be outlined; in the third section the two coupling procedures with TUBRNP will be discussed; the fourth section gives some information about the coupling with the oxygen redistribution model (OXIRED); in the fifth section, the revised version of PUREDI is tested by means of either specific or integral tests; the last section draws some conclusions.

2. Overview of the PUREDI model

Plutonium redistribution in nuclear fuel is important for the assessment of FBR rod performance since it can significantly affect fuel temperatures by changing the fuel thermal conductivity and the radial power profile. This effect is well known from post-irradiation examinations and out-of-pile experiments and is mainly caused by the following two mechanisms [1,2]:

(a) Solid state thermal diffusion
(b) Vapour transport by migrating pores and via cracks

The transport of plutonium by vapour migration via cracks was recognized as negligible for long-term operation [3,4], since cracks heal relatively fast during irradiation due to the high temperature in FBR fuel. The importance of vapour transport via pores had been also discussed in the past with different opinions and ideas [1,4,5,6,7]. According to Olander [1] and Clement and Finnis [4], the transport via pores should be of less importance compared to solid state diffusion, mainly because pore migration occurs immediately, inducing restructuring and formation of the central void. Therefore, this can have an effect on plutonium only during the first hours of irradiation, while solid state diffusion is a long-term process which produces a continuously growing redistribution effect. Only mechanism (a) is therefore considered in the Transuranus code.
On the basis of experimental radial profiles of plutonium in irradiated MOX fuels, Bober et al. [8] suggested the adoption of the standard equation of thermal diffusion:

$$\frac{\partial c_{Pu}}{\partial t} = -\nabla \cdot J_{Pu} \qquad (1)$$

$$J_{Pu} = -D_{U-Pu}\left(\nabla c_{Pu} + c_{Pu} c_U \frac{Q_{U-Pu}}{RT^2} \nabla T \right) \qquad (2)$$

where J_{Pu} is the vector flux of species per unit area and unit time; c_{Pu} and c_U ($\cong 1 - c_{Pu}$) are the molar fraction of plutonium and uranium oxides, respectively; R is the universal gas constant; T the absolute temperature; $Q_{U\text{-}Pu}$ is the effective molar heat of transport; and $D_{U\text{-}Pu}$ is the chemical interdiffusion coefficient.
Considering plutonium migration only along the radial direction (axial diffusion is neglected), Eqs. (1) and (2) can be rewritten in cylindrical coordinates as follows:

(3) $$\frac{\partial c_{Pu}}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(r J_{Pu})$$

(4) $$J_{Pu} = -D_{U-Pu}\left(\frac{\partial c_{Pu}}{\partial r} + c_{Pu}(1-c_{Pu})\frac{Q_{U-Pu}}{RT^2}\frac{\partial T}{\partial r}\right)$$

Since PUREDI deals only with redistribution (diffusion) of Pu, boundary conditions have to be chosen so as to ensure that no Pu is created or destroyed. This implies that the flux of Pu atoms at the inner (R_i) and outer (R_o) fuel surfaces is zero, as follows:

(5) $$J_{Pu}\big|_{r=R_i} = J_{Pu}\big|_{r=R_o} = 0$$

The solution of Eqs. (3) and (4) with boundary conditions (5) is obtained in the original PUREDI program according to the Lassmann's formulation of Bober's FBR redistribution model [9,10], which represents the starting point of the extension proposed in this report. The numerical solution is obtained by means of a finite difference scheme, which is summarized in Appendix A, under the following two assumptions:

(i) Linearization of Eq. (4) by imposing:

(6) $$c_{Pu}(1-c_{Pu}) \approx c_{Pu,n+1}(1-c_{Pu,n})$$

where n+1 indicates the actual and n the previous time step. In other words, the relative change of C_U is assumed to be small compared to the relative change of C_{Pu} in a time step.

(ii) Diffusion of Pu close to the outer fuel surface is low due to the low temperatures and low concentration gradient:

(7) $$c_{Pu,m} \approx c_{Pu,m-1}$$

where m indicates the last radial mesh point of the fuel. Hypothesis (ii) is adopted in order to avoid numerical oscillation in the solution caused by the boundary condition (5).
The numerical scheme of PUREDI (see Appendix A) has been recently revised, rewritten and extensively tested on the basis of a Monte Carlo analysis consisting of about 10^6 different cases. The results are not reported here for brevity.
The validity of the hypothesis (i) and (ii) has been verified by means of a code-to-code comparison with the finite element commercial software COMSOL Multiphysics [11]. The

solution provided by COMSOL does take into account the non-linearity of Eq. (4) and does not make use of the simplification given by Eq. (7). Figure 1 shows the comparison between PUREDI and COMSOL, as well as with the analytic solution provided by Clement [12]. As can be seen, both the above discussed hypotheses are valid, since PUREDI and COMSOL solutions are practically identical. The use of the analytic solution is only valid for very short times, because it has been derived by means of a first order integration of Eq. (3), and therefore cannot be used for the purposes of a fuel performance code.

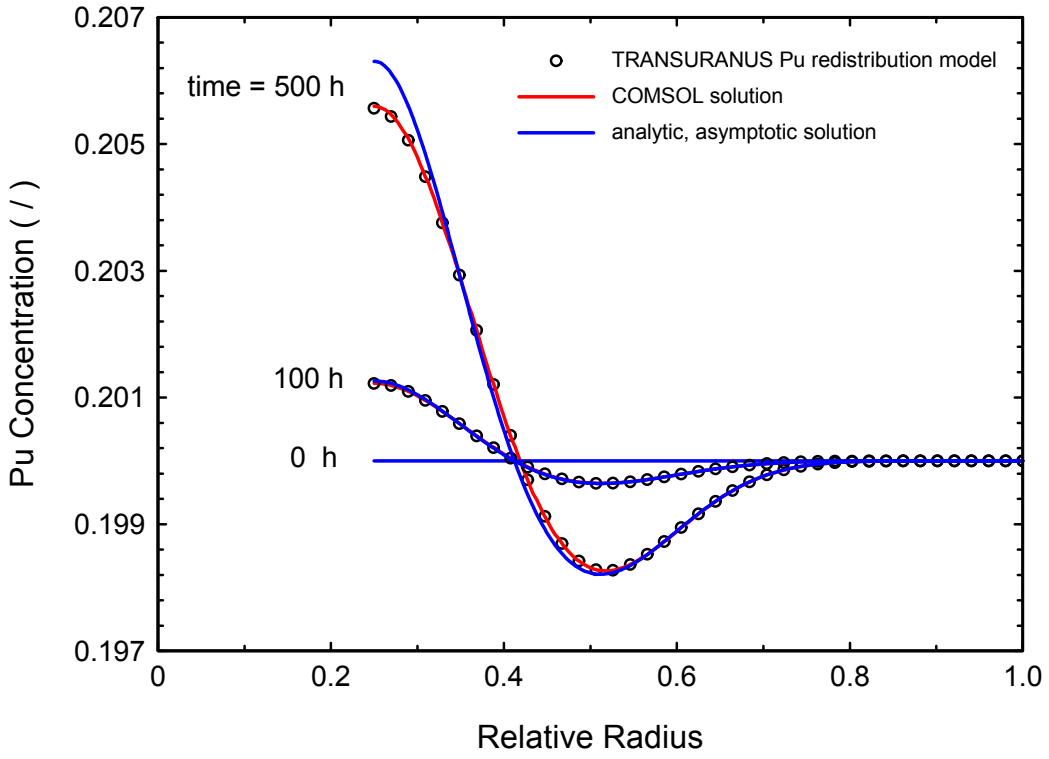

Figure 1: Radial distribution of Pu at different times obtained by means of PUREDI, COMSOL and the analytic solution [12].

3. Coupling PUREDI with TUBRNP

This chapter presents the extension of the PUREDI programme discussed in chapter 2 in order to take into account plutonium evolution due to burnup. In principle, an additional source term in the diffusion equation (1) (which is in general different for each plutonium isotope and radial dependent) should be considered. This would require a reformulation of the TUBRNP program and PUREDI, as well as of the Transuranus structure, because TUBRNP should be included in the implicit loop [10]. However, we can assume that: i) redistribution occurs much faster than burnup evolution in a given time step; ii) in FBR conditions, plutonium build-up or depletion (given by TUBRNP) has a very small radial gradient. It follows that the source term can be disregarded and the coupling can be simply performed by changing the initial value of plutonium distribution for PUREDI at each time step coming from TUBRNP. In particular, assumptions i) and ii) allow to keep PUREDI and TUBRNP separated within the Transuranus structure, and to solve the diffusion equation without the source term. More precisely, if the source term S = const, the solution of the diffusion equation can be written as $c_{Pu} = c_{Pu\_homo} + S \cdot t$ for small t, where c_{Pu_homo} is the solution of the homogeneous diffusion equation (i.e., Eq. (1)). $S \cdot t$ is a particular solution approximated at the first order (small t), and is considered in the present formulation as the initial value at each time step: $c_{Pu\_in} = c_{Pu\_homo}\ (t - \Delta t) + S \cdot \Delta T$. The influence of this approximation is discussed in Appendix C (Figure C.1), where the equation containing the source term is solved by means of COMSOL.

In order to couple PUREDI with TUBRNP we also need to calculate the redistributed concentrations of the five different plutonium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu) considered at each time step. We identified 2 different approaches to perform this task. Assuming that the transport phenomenon is the same for each plutonium isotope (the chemical behaviour is identical) and that neither $D_{U\text{-}Pu}$ nor $Q_{U\text{-}Pu}$ depend on plutonium concentration [8], we assume that the diffusion equation (Eq. (3)) can be written for the single plutonium isotope, as follows:

$$\frac{\partial c_{Pu,j}}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r J_{Pu,j}\right) \tag{8}$$

$$J_{Pu,j} = -D_{U-Pu}\left(\frac{\partial c_{Pu,j}}{\partial r} + c_{Pu,j}\left(1 - \sum_{j=1}^{5} c_{Pu,j}\right)\frac{Q_{U-Pu}}{RT^2}\frac{\partial T}{\partial r}\right) \tag{9}$$

where

(10) $$c_{Pu} = \sum_{j=1}^{5} c_{Pu,j} = c_{Pu-238} + c_{Pu-239} + c_{Pu-240} + c_{Pu-241} + c_{Pu-242}$$

The total flux of plutonium atoms is given by the sum of the isotopic fluxes, so that Eqs. (3) and (4) are still satisfied:

(11) $$J_{Pu} = \sum_{j=1}^{5} J_{Pu,j}$$

The previous approach is discussed in Appendix B.

Eqs. (8)-(11) consist in a non-linear system of five coupled partial differential equations, which would be difficult to solve. However, the non-linear term in Eq. (9) can be linearized in the same way as discussed in section 2:

(12) $$c_{Pu,j}\left(1 - \sum_{j=1}^{5} c_{Pu,j}\right) = c_{Pu,j}\left(1 - c_{Pu}\right) \approx c_{Pu,j,n+1}\left(1 - c_{Pu,n}\right)$$

Thanks to Eq. (12), each equation is not only simplified (linear), but, in a given time step, is independent on the concentration of the other isotopes. As a result, we obtained 5 diffusion equations for the 5 plutonium isotopes, which can be solved by means of the algorithm discussed in section 2 and Appendix A.

The main drawback of this approach is the computational time, which is increased at least by a factor of five compared to solving the diffusion equation for the total Pu. To improve PUREDI performance, we propose an alternative way to calculate the redistributed concentrations for each plutonium isotope. We simply assume that the concentrations of the different isotopes after redistribution can be obtained on the basis of their concentration calculated by TUBRNP in the same time step (i.e., before redistribution) as follows:

(13) $$c_{Pu,j}(r) = \frac{c_{Pu,j}^{BR}(r)}{c_{Pu}^{BR}(r)} \cdot c_{Pu}(r) = \alpha_j(r) \cdot c_{Pu}(r)$$

where

(14) $$\sum_{j=1}^{5} \alpha_j(r) = 1 \qquad \text{for all r}$$

where BR stands for "before redistribution". This approximation is discussed in Appendix C, where it is shown that Eqs. (3) and (4) are still fulfilled because of Eq. (14). On the contrary, the diffusion equation for each isotope (Eq. (8)) is not valid because an additional term depending on $\alpha_j(r)$ appears in the flux J_j. This term comes from the additional non-linearity introduced by Eq. (13) and does not assure mass conservation and boundary conditions for each plutonium isotope.

However, if the radial gradient of c_j is small compared to the second term of the right-hand side of Eq. (9), the two approaches are almost identical.

Both approaches[1] have been extensively tested as stand-alone programs, in which an additional depletion term (that modifies the initial plutonium concentration at each time step) is also introduced to simulate the TUBRNP effect. For the sake of brevity, only a part of this comparison will be shown.

Figure 2 shows the total plutonium concentration after 10000 hours with a constant depletion of $5 \cdot 10^{-3}$ %/hour for the different approaches. As expected the solutions are identical, because Eq. (3) is valid for both approaches.

An interesting comparison is shown in Figure 3, where different initial concentrations are adopted for the different plutonium isotopes (uniform depletion rate). Even if it cannot be seen in the graph, there are very small differences between the two approaches introduced by the term $\alpha_j(r)$. Actually, each isotope redistributes with a different importance having a different initial amount.

In order to visualize significant differences between the two approaches we have to introduce large radial gradients, for example by using a non-uniform depletion rate like in a LWR. As shown in Appendix C, this would cause a distortion in the plutonium radial profile which is of course non-physical.

However, the encountered discrepancies are always within the uncertainty associated to the measurements. Therefore, we consider for the implementation in Transuranus the simplified approach given by Eq. (13), so that the diffusion equation in PUREDI is solved only for the total plutonium concentration. This represents a good compromise between accuracy and computational costs.

[1] In the following, the solution labelled as "splitting" is the one obtained by solving five diffusion equations for the five Pu isotopes, while the solution labelled as "approximation" is obtained by means of Eq. (13).

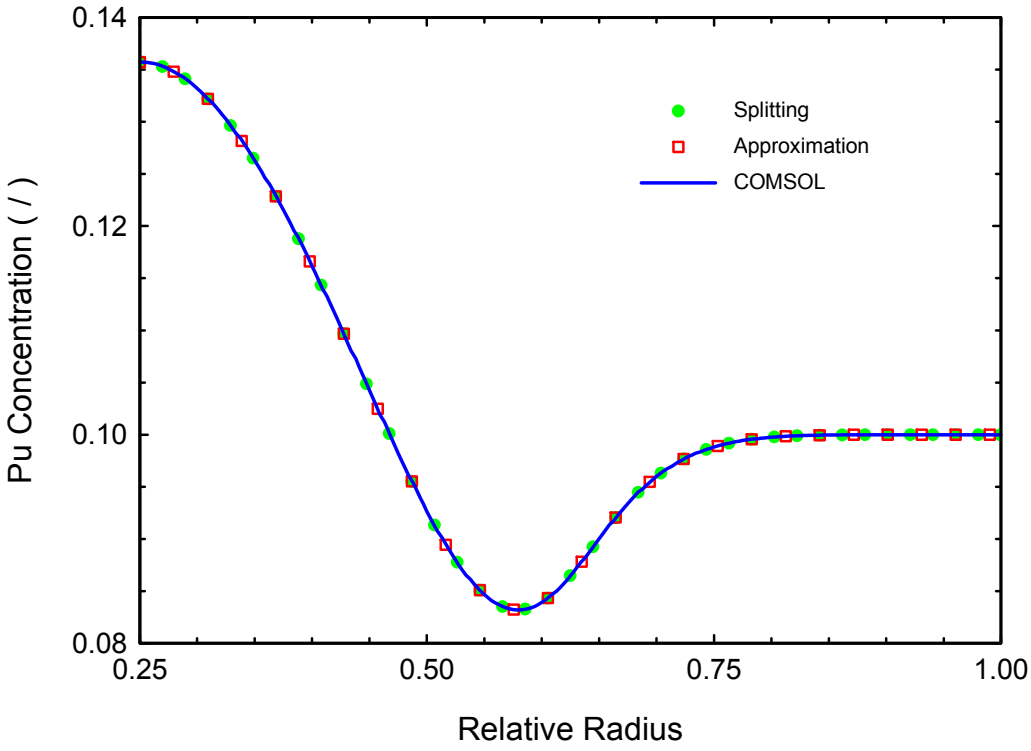

Figure 2: Comparison between COMSOL solution and the two different approaches proposed for PUREDI: total Plutonium concentration.

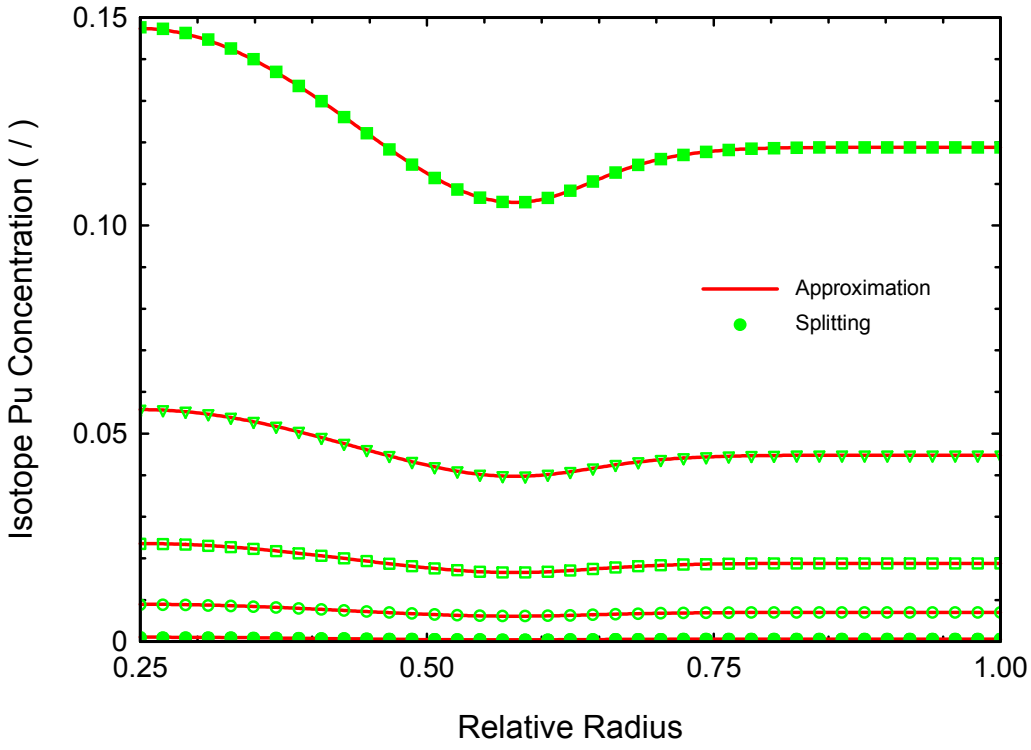

Figure 3: Comparison between the two different approaches proposed for PUREDI. Different initial concentrations for c_j and constant and uniform depletion rate.

4. Coupling PUREDI with OXIRED

The Transuranus oxygen redistribution programme (OXIRED) calculates the steady-state and transient radial oxygen-to-metal ratio (O/M) in a fuel rod [13]. The programme is based on the work of Sari and Schumacher [14] and predicts the O/M evolution according to the thermal diffusion equation:

$$(15) \qquad \frac{\partial c_{i,v}}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left\{ r D_O \left(\frac{\partial c_{i,v}}{\partial r} + c_{i,v} \frac{Q^*}{RT^2}\frac{\partial T}{\partial r} \right) \right\}$$

where $c_{i,v}$ is the concentration of oxygen vacancies/interstitials, Q* is the oxygen heat of transport and D_O is the diffusion coefficient.
The coupling with the plutonium redistribution in the fuel (q is the Pu molar fraction) is given by the dependence of the different parameters of Eq. (15):

a) the heat of transport (for oxygen vacancies for hypostoichiometric fuels) depends on the plutonium valence V_{Pu} in the fuel according to the following correlation [14]:

$$(16) \qquad Q^* = -9.45 \cdot 10^5 + 5.66 \cdot 10^5 V_{Pu} - 8.5 \cdot 10^4 V_{Pu}^2$$

where

$$(17) \qquad V_{Pu} = 4 + \frac{2(O/M - 2)}{q}$$

b) the diffusion coefficient depends on the Plutonium content and the correlation adopted for D_O has been recently updated [15].

5. Tests of the PUREDI model

The revised PUREDI model of the Transuranus code has been extensively tested by means of specific tests concerning the stand-alone version of PUREDI and by means of integral tests concerning fuel pins irradiated in fast reactor. Besides the modifications concerning the coupling with TUBRNP (discussed in this report), also the plutonium interdiffusion coefficient has been revised. A proposal for $D_{U\text{-}Pu}$ was made in Ref. [15], in order to take into account the dependence of the diffusion mechanism on the oxygen-to-metal ratio. The results of the integral test will be shown with the new correlation for the diffusion coefficient.

5.1 Specific tests

Specific tests consisted in the analysis of the stand-alone model of PUREDI. This has been carried out by means of a Monte Carlo Test. The program runs 100 cases with more than 200.000 calls to PUREDI and PUIMPL (see Appendix A). This test was performed to check: 1) if the revised and the old version of PUIMPL give the same results; 2) the performance of the revised PUREDI package. The comparison between both versions shows only few minor differences due to round off errors. As concerns the performance of the new PUREDI package an increase in the computational time of a factor of 3 has been found probably due to the implementation of the new diffusion coefficient.

5.2 Application of the revised PUREDI model to irradiations

Two cases from the SUPERFACT experiment have been analysed. They consist in two fuel rods with MOX fuel containing few percents of minor actinides irradiated in the fast reactor of Phenix. They are labelled as SF-2%Np and SF-2%Am. The first one experienced higher plutonium redistribution due to the high power level and temperatures reached during irradiation. EPMA measurements were carried out at the ITU. The comparison between Transuranus with the revised PUREDI model and the experimental data is shown in Figures 4 and 5. A good agreement has been found and the discrepancies are within the uncertainties of the measurements.

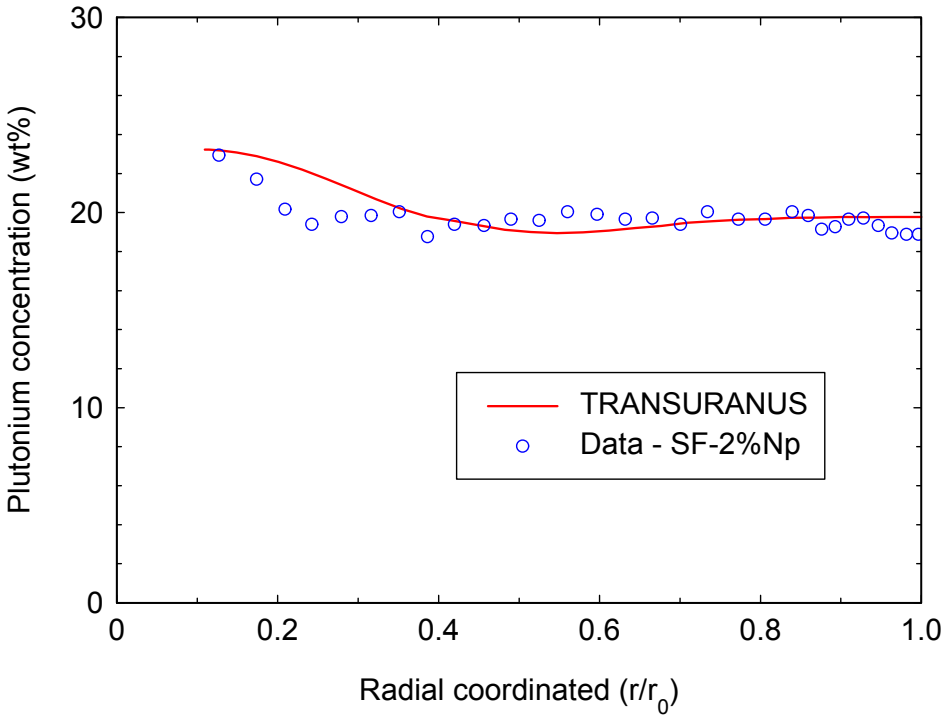

Figure 4: Comparison between Transuranus and EPMA measurements of Plutonium redistribution for the fuel pin SF-2%Np.

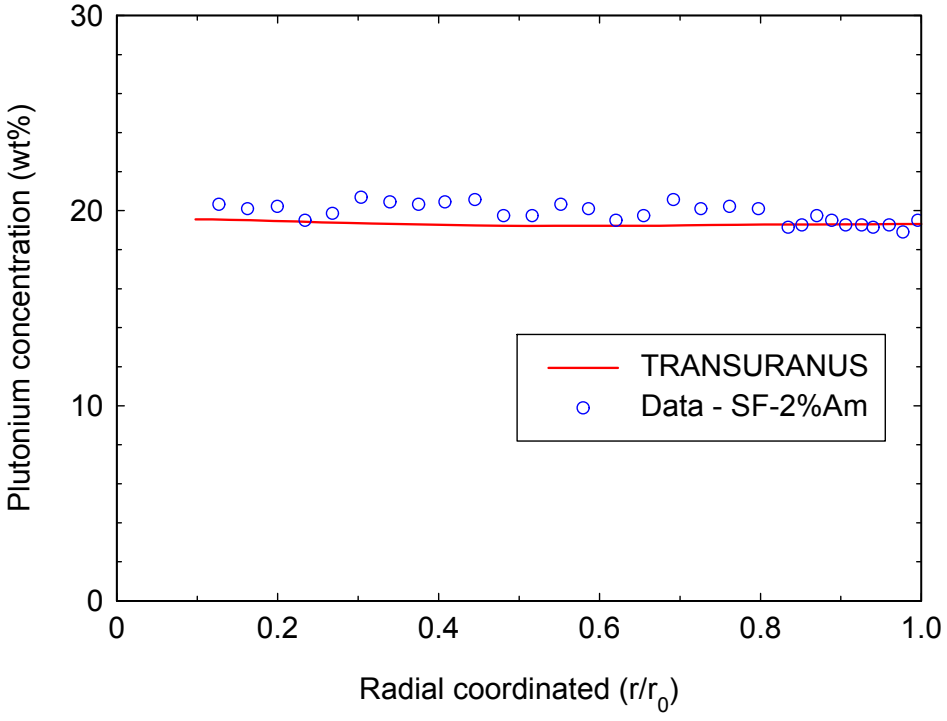

Figure 5: Comparison between Transuranus and EPMA measurements of Plutonium redistribution for the fuel pin SF-2%Am.

5.3 Incompatibility of the PUREDI model with LWR conditions

It is important to note that the hypothesis (ii) discussed in section 2 (required by equation (5)) is obviously not applicable to large gradients of the local Pu concentration at the periphery of the fuel. Figure 6 illustrates the situation for the OSIRIS H09 rod of the FUMEX-III project (irradiation in PWR, radially averaged burn-up in the mid-rod axial position: 44 MWd/kgHM): The assumption $c_{Pu.m} \approx c_{Pu,m-1}$ (Figure 6a) results in a distortion of the local burn-up (Figure 6b). Furthermore, due to the formation of the high burn-up structure (HBS) at the periphery of the fuel, the modified local burn-up has a significant impact on the resulting local Xe concentration (Figure 6c). Tests have shown that this problem can not be solved by increasing the number of radial nodes, because the gradient of the Pu concentration is increasing towards the surface of the fuel.

As such a situation occurs for any UO_2 or MOX fuel irradiated in a thermal-neutron environment - typically at intermediate burn-up - the PUREDI model should in general not be applied for simulating irradiations in light-water reactors (LWR). The extension of the Transuranus code in this case foresees an automatic switch off of the PUREDI model (IPURE=0, and a warning message is given to the user).

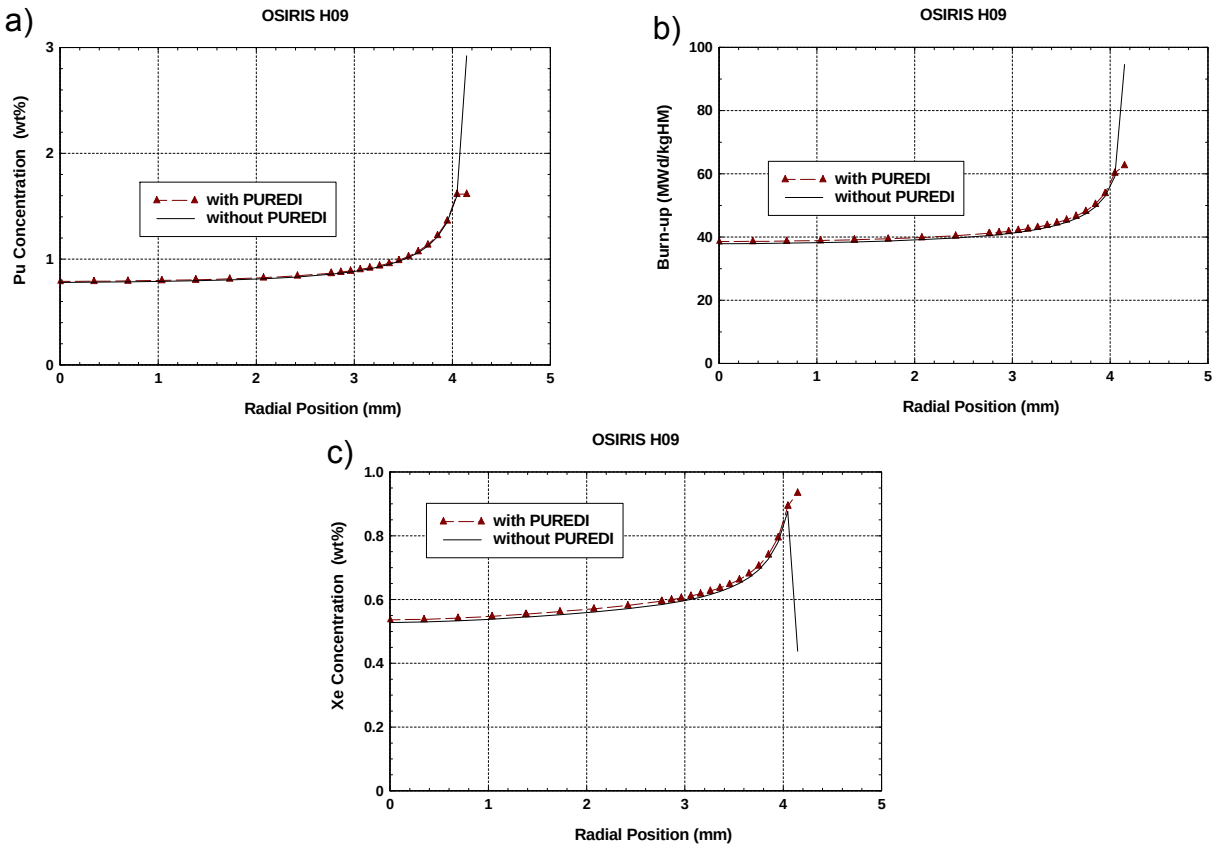

Figure 6: Comparison of simulated local Pu concentration, the local burn-up and the local Xe concentration for a priority case the FUMEX-III project.

6. Conclusions and perspectives

In this report the revised version of the PUREDI programme for the calculation of the Plutonium redistribution in fast reactor fuels has been discussed. The programme has been completely rewritten for the fortran 95 version of the code. The numerical structure has been revised and extensively tested by means of a Monte Carlo test program.

In the context of calculating plutonium redistribution for fast reactor fuel rods simulations, the new PUREDI version has been coupled with TUBRNP by means of a simplified approach which preserves computational time maintaining the same accuracy. In particular, the concentrations of the different plutonium isotopes are calculated starting from the total plutonium concentration (calculated by PUREDI) on the basis of the isotopic ratio obtained from TUBRNP in the same time step but before redistribution. The limitations of this approach have also been discussed. Finally, the programme has been verified on the basis of integral tests (two rods of the SUPERFACT experiment) showing a good agreement with the experimental data.

The revised version of PUREDI presented in this report can be easily extended for the calculation of actinides redistribution (Am, Np, etc.) once their transport behaviour in UO_2 matrix will be assessed. PUREDI represents a reliable program to deal also with minor actinides fuels, which are foreseen to be adopted in the future in order to reduce the inventory and radiotoxicity of fuel cycle waste.

Appendix A: Implementation of the Transuranus PUREDI model

In this Appendix the finite difference algorithm adopted to solve the diffusion equation is described. The algorithm was developed in [9,10] according to Lassmann's approach to Bober's redistribution model and the equations are herein rewritten in a better understandable form. A small modification to original Lassmann's algorithm is proposed (i.e., for the last equation) in order to avoid oscillations in the numerical solution when diffusion is important at the outer fuel surface.

The PUREDI model implemented in Transuranus consists of 3 subroutines according to the following scheme:

PUREDI → PUIMPL → FDIAG

where PUREDI is the driver, PUIMPL includes the finite difference scheme and FDIAG solves a pentadiagonal system of equations. This role was played in the previous PUREDI model by 3 subroutines (i.e., FDIAG, FDIAGL and FDIAGZ) which have been now incorporated into a single routine, that is FDIAG.

The finite difference approach and the relative equations are derived below. The discretization in time will take the index n in the following (n + 1 indicates the current time step). The fuel is divided into m radial zones which need not to be equidistant. In the following, a superscript (i) indicates a radial zone whereas a subscript indicates the value at the node r_i (see Figure A.1). For simplicity, the subscript Pu in the definition of the physical quantities in section 2 is omitted (i.e., $c = c_{Pu}$, $D = D_{Pu}$, $J = J_{Pu}$, etc.).

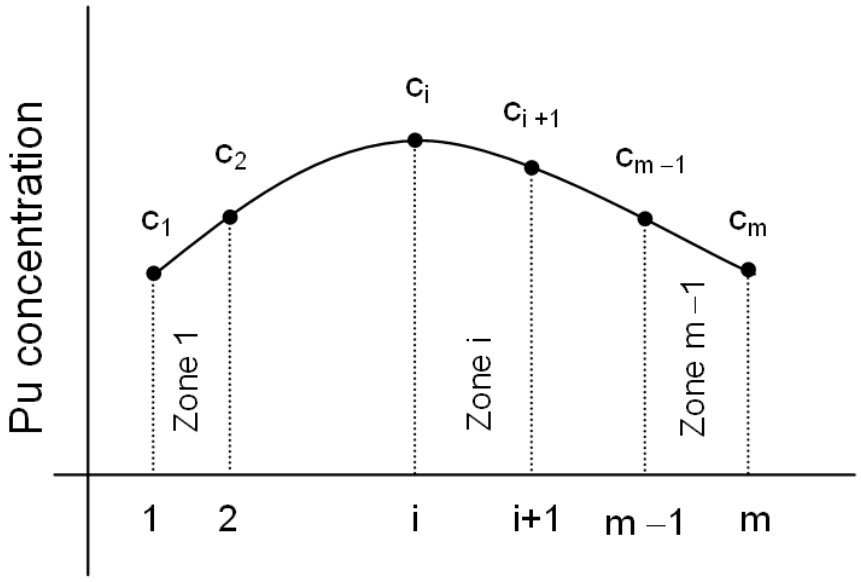

Figure A.1: Discretization scheme of PUREDI model.

For each zone the balance equation is written:

(A.1) $$w^{(1)}\dot{c}_1 + \left(1 - w^{(1)}\right)\dot{c}_2 = -\frac{2r_2J_2}{r_2^2 - r_1^2}$$ Zone 1

(A.2) $$w^{(i)}\dot{c}_i + \left(1 - w^{(i)}\right)\dot{c}_{i+1} = -\frac{2\left(r_{i+1}J_{i+1} - r_iJ_i\right)}{r_{i+1}^2 - r_i^2}$$ Zone 2….m –2

(A.3) $$w^{(m-1)}\dot{c}_{m-1} + \left(1 - w^{(m-1)}\right)\dot{c}_m = +\frac{2r_{m-1}J_{m-1}}{r_m^2 - r_{m-1}^2}$$ Zone m –1

where w is an area weighting factor defined as:

(A.4) $$w^{(i)} = \frac{r_{i+1} + 2r_i}{3\left(r_{i+1} + r_i\right)}$$

and the time derivative of plutonium concentration is discretized as follows:

(A.5) $$\dot{c}_i = \frac{c_{i,n+1} - c_{i,n}}{\Delta t}$$

The equation system to be solved consist of m – 1 equations for m unknowns, so that an additional equation is needed. To close the system, we adopt the hypothesis discussed in section 2, namely $c_m = c_{m-1}$.

Concerning the term J_i, defined by Eq. (4), its discretization requires the definition of the radial derivative of c:

(A.6) $$\left.\frac{\partial c}{\partial r}\right|_i \approx \frac{1}{2}\frac{c_{i+1} - c_i}{r_{i+1} - r_i} + \frac{1}{2}\frac{c_i - c_{i-1}}{r_i - r_{i-1}}$$

The subscript n is omitted since c_i are taken at time n+1 (i.e. are unknown). This is a fully implicit formulation. A comparison between an implicit and an explicit scheme was also performed showing a perfect agreement. However, since the explicit solution did not result in significant reductions of the computational costs, only the fully implicit treatment was incorporated into the Transuranus code.

Now, we can define:

(A.7) $$f_{i+1} = \frac{1}{2}\frac{1}{r_{i+1} - r_i}\ ;\ \ f_i = \frac{1}{2}\frac{1}{r_i - r_{i-1}}$$

and obtain:

(A.8) $$\left.\frac{\partial c}{\partial r}\right|_i \approx c_{i+1}\cdot f_{i+1} + c_i\left(f_i - f_{i+1}\right) - c_{i-1}\cdot f_i$$

According to the linearization of Eq.(6) in section 2 we define:

(A.9) $$g_i = \left(1 - c_{i,n}\right)\frac{Q}{RT^2}\frac{\partial T}{\partial r}$$

where all values are known, since they are defined at the previous time step and taken at r = r_i. So, we can rewrite the term J_i as follows according to Eqs. (A.6)-(A.9):

(A.10) $$J_i = c_{i+1}\cdot f_{i+1} + c_i\left(f_i - f_{i+1}\right) - c_{i-1}\cdot f_i + c_i\cdot g_i$$

In addition, we define the following terms:

(A.11) $$h_{i+1} = \frac{2r_{i+1}}{r_{i+1}^2 - r_i^2}D_{i+1}\Delta t\ ; \qquad h_i^* = \frac{2r_i}{r_{i+1}^2 - r_i^2}D_i\Delta t$$

Now, we can rewrite Eqs. (A.1) and (A.2) making use of the previous definitions. Starting from Zone 1:

(A.12) $$\begin{aligned} &c_1\left\{w^{(1)} + h_2 f_2\right\} + \\ &c_2\left\{\left(1 - w^{(1)}\right) - h_2\left(f_2 - f_3 + g_2\right)\right\} + \\ &c_3\left\{-h_2 f_3\right\} = \\ &w^{(1)}c_{1,n} + \left(1 - w^{(1)}\right)c_{2,n} \end{aligned} \qquad \text{Zone 1}$$

In a similar way, we proceed for Zone 2…m – 2:

(A.13) $$\begin{aligned} &c_{i-1}\left\{-h_i^* f_i\right\} + \\ &c_i\left\{w^{(i)} + h_{i+1}f_{i+1} + h_i^*\left(-f_{i+1} + f_i + g_i\right)\right\} + \\ &c_{i+1}\left\{\left(1 - w^{(i)}\right) - h_{i+1}\left(-f_{i+2} + f_{i+1} + g_{i+1}\right) + h_i^* f_{i+1}\right\} + \\ &c_{i+2}\left\{-h_{i+1}f_{i+2}\right\} = \\ &w^{(i)}c_{i,n} + \left(1 - w^{(i)}\right)c_{i+1,n} \end{aligned} \qquad \text{Zone 2…m – 2}$$

As far as the equation for the last zone is concerned, the original Lassmann's algorithm starts from Eq. (A.3) and applies the hypothesis discussed in section 2 ($c_{m-1} = c_m$). As shown in Figure A.2, this approach can lead to oscillation in the solution if diffusion is important at the outer fuel surface, because Eq. (A.3) has been derived assuming $J_m = 0$, which is not consistent with the hypothesis $c_{m-1} = c_m$.

To overcome this problem we propose to start from Eq. (A.13) also for the last zone. To proceed, we need to redefine the concentration gradient at the outer fuel surface:

$$\left.\frac{\partial c}{\partial r}\right|_m = \frac{c_m - c_{m-1}}{r_m - r_{m-1}} = (c_m - c_{m-1}) f_m \qquad \text{(A.14)}$$

where

$$f_m = \frac{1}{r_m - r_{m-1}} \qquad \text{(A.15)}$$

Hence, the equation for the last zone is:

$$\begin{aligned} & c_{m-2}\left\{-h^*_{m-1} f_{m-1}\right\} + \\ & c_{m-1}\left\{w^{(m-1)} + h_m f_m + h^*_{m-1}\left(-f_m + f_{m-1} + g_{m-1}\right)\right\} + \\ & c_m\left\{\left(1 - w^{(m-1)}\right) - h_m\left(f_m + g_m\right) + h^*_{m-1} f_m\right\} = \\ & w^{(i)} c_{m-1,n} + \left(1 - w^{(i)}\right) c_{m,n} \end{aligned} \qquad \text{Zone } m-1 \qquad \text{(A.16)}$$

The final equation is represented by the hypothesis discussed in section 2: $c_m = c_{m-1}$.

The equation system given by Eqs. (A.12)-(A.16) is implemented in the subroutine PUIMPL and is solved by the FDIAG subroutine. A comparison between the two algorithms is given in Figure A.2.

Since the fully implicit scheme guarantees numerical stability even for extremely large time steps, the time step length, which controls the plutonium redistribution, has to be limited because of the linearization adopted in Eq. (6). Numerous tests proved that the time step criterion

$$\Delta t \le 0.01 \cdot \max\left\{c_{i,n+1} - c_{i,n}\right\} \qquad \text{(A.16)}$$

is a good compromise between accuracy and computational cost.

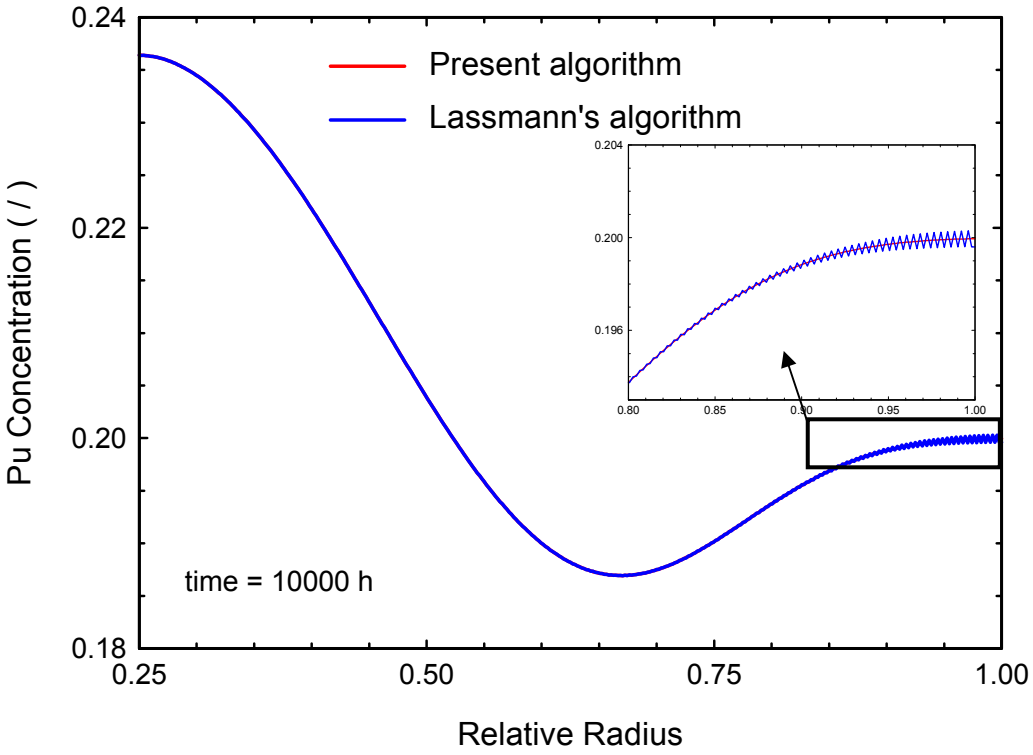

Figure A.2: Comparison between Lassmann's algorithm and the present one proposed for PUREDI. The diffusion coefficient derived in [15] is used for the comparison. This correlation returns higher values of $D_{U\text{-}Pu}$ (even a factor of 15 at low temperatures according to the oxygen to metal ratio) with respect to that suggested in Ref. [8].

Appendix B: Validity of the diffusion equation for each plutonium isotope

In this appendix, the problems given by Eq. (8) and (9) will be discussed. For the sake of simplicity, we rewrite again Eqs. (3) and (4) omitting the subscript Pu:

$$\frac{\partial c}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(rJ) \tag{B.1}$$

$$J = -D\left(\frac{\partial c}{\partial r} + c(1-c)\frac{Q}{RT^2}\frac{\partial T}{\partial r}\right) \tag{B.2}$$

The total plutonium concentration is given by the sum of the concentrations of the different isotopes:

$$c = \sum_{j=1}^{5} c_j \tag{B.3}$$

Since the non-linear term is simplified (as shown in section 2) to get the solution in the PUREDI model, it can be considered as known. This approximation has been extensively tested for the PUREDI model, and is consistent as far as the time step is small. It follows that Eq. (B.1) is a linear differential equation. Thanks to this property, we can define a linear operator L (γ), which associates to a function γ the diffusion equation, as follows:

$$L(\gamma) := \frac{\partial \gamma}{\partial t} - \frac{1}{r}\frac{\partial}{\partial r}(rJ(\gamma)) = 0 \tag{B.4}$$

From Eq. (B.3), we can write:

$$L(c) = \sum_{j=1}^{5} L(c_i) \tag{B.5}$$

Since c_j must have the same properties as c (mass balance and boundary conditions – see below), it is reasonable to assume that the concentration of each plutonium isotope satisfies the diffusion equation:

$$L(c_j) = \frac{\partial c_j}{\partial t} - \frac{1}{r}\frac{\partial}{\partial r}(rJ(c_j)) = 0 \tag{B.6}$$

Now, some features of this approach will be discussed. Substituting Eq. (B.3) into (B.2) and then in (B.1), we get:

$$\frac{\partial\left(\sum_{j=1}^{5} c_j\right)}{\partial t} = -\frac{1}{r}\frac{\partial}{\partial r}\left(rD\left(\frac{\partial\left(\sum_{j=1}^{5} c_j\right)}{\partial r} + \left(\sum_{j=1}^{5} c_j\right)\left(1-\left(\sum_{j=1}^{5} c_j\right)\right)\frac{Q}{RT^2}\frac{\partial T}{\partial r}\right)\right) \tag{B.7}$$

So, we obtain:

$$\sum_{j=1}^{5}\frac{\partial c_j}{\partial t} = -\frac{1}{r}\frac{\partial}{\partial r}\left(rD\left(\sum_{j=1}^{5}\frac{\partial c_j}{\partial r} + \left(\sum_{j=1}^{5} c_j\right)(1-c)\frac{Q}{RT^2}\frac{\partial T}{\partial r}\right)\right) \tag{B.8}$$

and hence:

$$\sum_{j=1}^{5}\left\{ \frac{\partial c_j}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left(rD\left(\frac{\partial c_j}{\partial r} + c_j(1-c)\frac{Q}{RT^2}\frac{\partial T}{\partial r}\right)\right)\right\} = 0 \tag{B.9}$$

We can now define J_j as the flux of the j plutonium isotope:

$$J_j = -D\left(\frac{\partial c_j}{\partial r} + c_j(1-c)\frac{Q}{RT^2}\frac{\partial T}{\partial r}\right) \tag{B.10}$$

and hence

$$\sum_{j=1}^{5}\left\{ \frac{\partial c_j}{\partial t} - \frac{1}{r}\frac{\partial}{\partial r}\left(rJ_j\right)\right\} = 0 \tag{B.11}$$

Eq. (B.11) is the same as Eq. (B.5), and represents the verification of the linearity of the diffusion equation under consideration.

Now, we multiply both sides of Eq. (B.11) by r and integrate between R_i and R_o (inner and outer fuel radius, respectively). Since the integral is a linear operator, we can write:

$$\sum_{j=1}^{5}\left\{ \frac{\partial}{\partial t}\int_{R_i}^{R_o} rc_j dr - \left(R_o J_j\Big|_{r=R_o} - R_i J_j\Big|_{r=R_i}\right)\right\} = 0 \tag{B.12}$$

The integral in the first term of Eq. (B.12) represents the mass of each plutonium isotope. Since mass conservation must be assured for each isotope, we write:

(B.13) $$\frac{\partial}{\partial t}\int_{R_i}^{R_o} r c_j dr = 0 \qquad \text{for } j = 1,\ldots.5$$

The same argument can be applied to the second term, which represents the boundary conditions. Since no flux of atoms can occur at boundaries the second term is equal to zero for each j:

(B.14) $$J_j\Big|_{r=R_i} = 0 \qquad J_j\Big|_{r=R_o} = 0$$

Appendix C: Alternative approach to couple PUREDI with TUBRNP

The alternative approach (indicated as "approximation" in the following) proposed for the coupling between PUREDI and TUBRNP consists in splitting the total redistributed plutonium concentration on the basis of the values given by TUBRNP in the same time step, but before redistribution (the subscript Pu is omitted):

$$c_j = \frac{c_j^{BR}}{c^{BR}} \cdot c = \alpha_j(r) \cdot c \tag{C.1}$$

$$c = \sum_{j=1}^{5} c_j \tag{C.2}$$

We now verify that this approach assures the same total flux of atoms. Substituting Eqs. (C.1) and (C.2) into Eq. (B.10) and omitting the time index subscript, we get:

$$J_j = -D\left(\frac{\partial(\alpha_j(r) \cdot c)}{\partial r} + \alpha_j(r) \cdot c(1-c) \frac{Q}{RT^2} \frac{\partial T}{\partial r} \right) \tag{C.3}$$

Expanding the radial derivative, we can write:

$$\begin{aligned} J_j = -D\left(c \frac{\partial \alpha_j(r)}{\partial r} + \alpha_j(r) \left\{ \frac{\partial c}{\partial r} + c(1-c) \frac{Q}{RT^2} \frac{\partial T}{\partial r} \right\} \right) = \\ -Dc \frac{\partial \alpha_j(r)}{\partial r} + \alpha_j(r) \cdot J \end{aligned} \tag{C.4}$$

From Eq. (11) and Eq. (14), we can easily verify that the total flux of atoms is given by the sum of the different fluxes:

$$J = \sum_{j=1}^{5} J_j = -Dc \frac{\partial \sum_{j=1}^{5} \alpha_j(r)}{\partial r} + \sum_{j=1}^{5} \alpha_j(r) \cdot J_j = J \tag{C.4}$$

For this reason, the calculated total plutonium concentration is the same between the two approaches. The same cannot be said for the concentrations of the different isotopes. In fact, if we look at Eq. (C.4), we can identify an additional term proportional to the gradient of $\alpha_j(r)$ which comes directly from the non-linearity introduced by Eq. (C.1). Because of this term, the boundary conditions (see Eq. (B.14)) are not satisfied because this term is generally different

from zero at boundaries. The same is valid for the mass balance. However, the gradient of $\alpha_j(r)$ is very small compared to the temperature gradient, and to see some differences between the 2 approaches, strong gradient concentrations must be artificially introduced. For example, two cases with non-uniform depletion rate will be shown: 1) same initial concentration of c_j with different depletion rate non-uniform along r; 2) different initial concentration of c_j with strong gradient in the depletion radial profile.

The results are shown in Figures C.1 and C.2. In the first case, some discrepancies can be identified near the pellet centre, where the overestimation of the concentration for the first isotope is counterbalanced by the underestimation of the others.

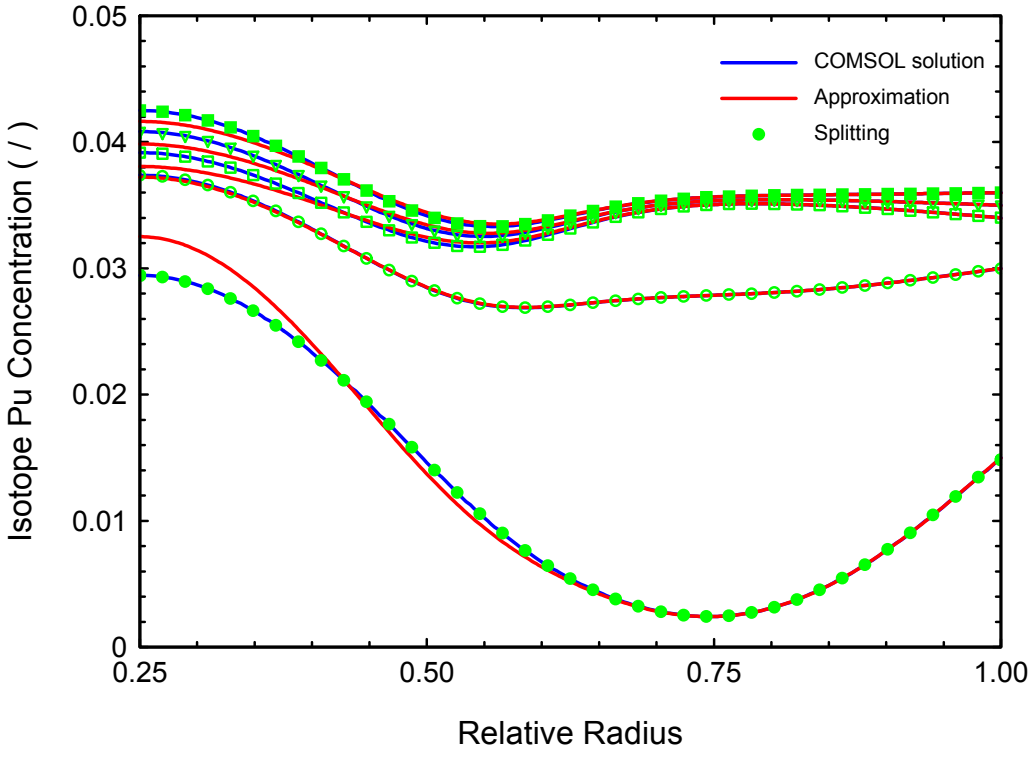

Figure C.1: Comparison between the two different approaches proposed for PUREDI and the numerical solution obtained by means of COMSOL. Same initial concentration of c_j but different depletion rate.

In this picture, also the COMSOL solution is given in order to assess the importance of the assumption related to the source term (see chapter 3). As can be seen, the effect of disregarding the source term is negligible compared to the error introduced by the gradient of $\alpha_j(r)$. The second case shows the distortion in the radial profile introduced by the additional term in Eq. (C.4), which is non-physical.

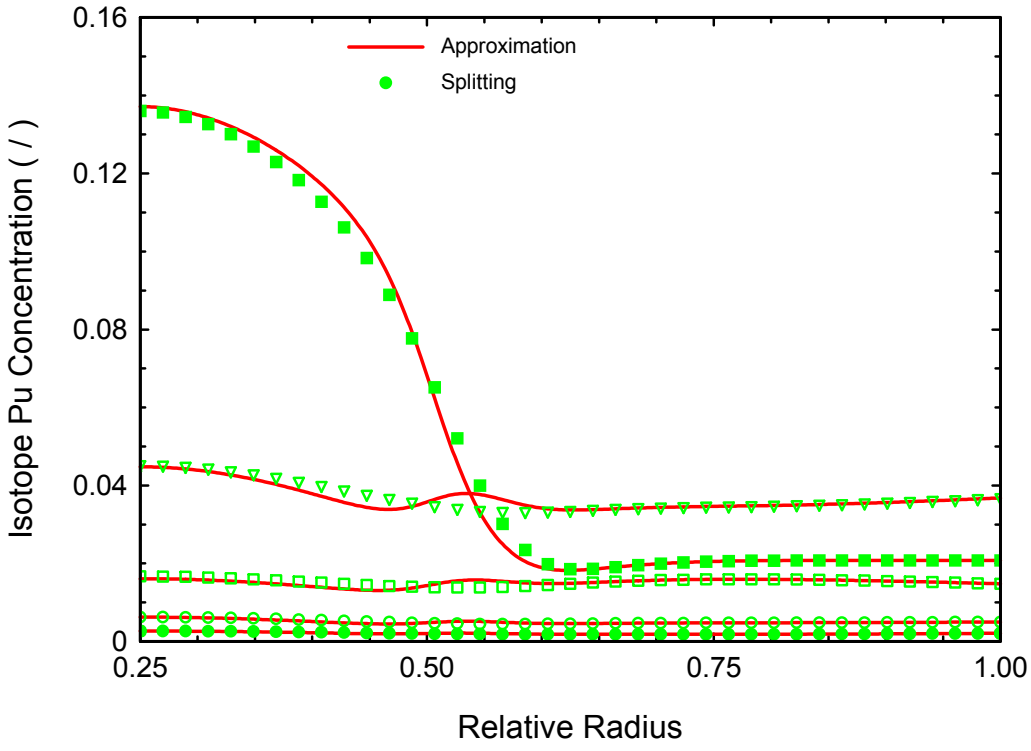

Figure C.2: Comparison between the two different approaches proposed for PUREDI. Different initial concentration of c_j with strong radial gradient in the depletion rate.

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Abstract
The Transuranus PUREDI model calculates the plutonium redistribution across the fuel pellet due to thermal diffusion. This phenomenon is particularly significant in oxide fuels for fast breeder reactors (FBR) where temperatures and temperature gradients are extremely high. The first version of PUREDI was developed and implemented as a stand-alone model. Since plutonium redistribution due to transport can lead to significant modifications of the radial power profile, a coupling of the PUREDI model with TUBRNP (recently extended for FBRs) is needed to correctly predict the fuel temperature. To this purpose, a revision of PUREDI has been proposed and the main features will be outlined in this report.

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